

## AMENDMENTS TO THE CLAIMS

The following listing of claims will replace all prior versions, and listings, of claims in the application.

### **Listing of the Claims:**

1.-43. (Canceled)

44. (Currently amended) An article comprising a substrate having at least one main surface coated with a multi-layer anti-reflection stack, wherein the multi-layer anti-reflection stack comprises, in the order indicated starting from the substrate:

(a) a high index (HI) layer, having a refractive index  $n_D^{25}$  of 1.50 to 2.00 and resulting from the hardening of a first hardenable composition and comprising an organic-inorganic hybrid matrix resulting from the hydrolysis and condensation of at least one precursor compound bearing an epoxy or (meth)acryloxy group and at least two functions hydrolysable to silanol groups, within which at least one colloidal metal oxide or at least one colloidal chalcogenide or a mixture of these compounds is dispersed in the form of particles from 1 to 100 nm in diameter, and directly on this high index layer (HI); ~~and~~

(b) a low index (LI) layer, having a refractive index  $n_D^{25}$  ranging from 1.38 to 1.44 obtained by deposition and hardening of a second hardenable composition and comprising the product of hydrolysis and condensation of:

(i) at least one precursor compound (I) comprising four hydrolysable functions per molecule of formula



in which the groups W, identical or different, are hydrolysable groups and provided that the groups W do not all represent at the same time a hydrogen atom; and

(ii) at least one precursor silane (II) bearing at least one fluorinated group and comprising at least two hydrolysable groups per molecule, said second composition comprising at least 10% by mass of fluorine in its theoretical dry extract (TDE), and the molar ratio  $[[\text{I}/\text{I} + \text{II}]] \frac{\text{I}}{\text{I} + \text{II}}$  of the

precursor compound (I) to the sum of the precursor compound (I) + precursor silane (II) of the second composition being greater than 80%.

45. (Previously presented) The article of claim 44, wherein a main surface of the substrate is coated with an anti-abrasion layer or a layer of a primer coating and a layer of an anti-abrasion coating, the anti-reflection stack being deposited onto the anti-abrasion coating.

46. (Previously presented) The article of claim 44, wherein, in addition, silica ( $\text{SiO}_2$ ) is dispersed in the matrix of the high index layer.

47. (Currently amended) The article of claim 44, wherein the colloidal metal oxides and chalcogenides dispersed in the matrix of the high index layer is  $\text{TiO}_2$ ,  $\text{ZnO}$ ,  $\text{ZnS}$ ,  $\text{ZnTe}$ ,  $\text{CdS}$ ,  $\text{CdSe}$ ,  $\text{IrO}_2$ ,  $\text{WO}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{FeTiO}_3$ ,  $\text{BaTi}_4\text{O}_9$ ,  $\text{SrTiO}_3$ ,  $\text{ZrTiO}_4$ ,  $\text{MoO}_3$ ,  $[\text{CO}_3\text{O}_4]$ ,  $\text{Co}_2\text{O}_4$ ,  $\text{SnO}_2$ , bismuth-based ternary oxide,  $\text{MoS}_2$ ,  $\text{RuO}_2$ ,  $\text{Sb}_2\text{O}_4$ ,  ~~$\text{BaTi}_4\text{O}_9$~~ ,  $\text{MgO}$ ,  $\text{CaTiO}_3$ ,  $\text{V}_2\text{O}_5$ ,  $\text{Mn}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{Nb}_2\text{O}_5$ , or  $\text{RuS}_2$ .

48. (Previously presented) The article of claim 44, wherein the particles of metal oxide dispersed in the matrix of the high index layer are constituted by a composite titanium oxide in the form of rutile.

49. (Previously presented) The article of claim 47, wherein the mineral particles dispersed in the organic-inorganic hybrid matrix of the high index layer (HI) have a composite structure based on  $\text{TiO}_2$ ,  $\text{SnO}_2$ ,  $\text{ZrO}_2$  and  $\text{SiO}_2$ .

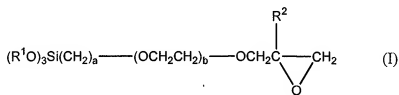
50. (Previously presented) The article of claim 44, wherein at least 60% by mass of the theoretical dry extract (TDE) of the low index layer are derived from the precursor compound (I).

51. (Previously presented) The article of claim 50, wherein at least 65% by mass of the theoretical dry extract (TDE) of the low index layer are derived from the precursor compound (I).

52. (Previously presented) The article of claim 51, wherein at least 70% by mass of the theoretical dry extract (TDE) of the low index layer are derived from the precursor compound (I).

53. (Currently amended) The article of claim 44, wherein the molar ratio  $\frac{[I]}{[I+II]}$  of the precursor compound (I) to the sum of the precursor compound (I) + precursor silane (II) is at least 85%.
54. (Currently amended) The article of claim 53, wherein the molar ratio  $\frac{[I]}{[I+II]}$  of the precursor compound (I) to the sum of the precursor compound (I) + precursor silane (II) is at least 90%.
55. (Currently amended) The article of claim 54, wherein the molar ratio  $\frac{[I]}{[I+II]}$  of the precursor compound (I) to the sum of the precursor compound (I) + precursor silane (II) is at least 95%.
56. (Previously presented) The article of claim 44, wherein the hydrolysable groups W represent an OR, Cl or H group, R being alkyl.
57. (Previously presented) The article of claim 44, wherein the hardenable composition of the low index layer (LI) comprises a tri- or dialkoxysilane different from the silanes of the precursor compound (I) of formula  $Si(W)_4$  and from the precursor fluorosilane (II) in a proportion by weight not exceeding 10% of the total weight of the silanes present in said composition.
58. (Currently amended) The article of claim 44, wherein the hardenable composition of the low index layer (LI) does not comprise ~~[[s]] only the~~ silanes ~~[[of]] other than~~ the precursor compound (I) and the precursor fluorosilane (II).
59. (Currently amended) The article of claim 44, wherein the anti-reflection stack ~~comprises only~~ consists of a high index layer (HI) coated with a low index layer (LI).
60. (Previously presented) The article of claim 44, wherein the anti-reflection stack comprises at least three superimposed layers, starting from the substrate, a medium index layer (MI), a high index layer (HI) and a low index layer (LI), respectively, the medium index layer (MI) having a refractive index  $n_D^{25}$  of 1.45 to 1.80.
61. (Previously presented) The article of claim 44, wherein the layer of material of high refractive index (HI) has a refractive index greater than 1.7.

62. (Previously presented) The article of claim 61, wherein the layer of material of high refractive index (HI) has a refractive index ranging from 1.72 to 1.82.
63. (Previously presented) The article of claim 62, wherein the layer of material of high refractive index (HI) has a refractive index of 1.77.
64. (Previously presented) The article of claim 44, wherein the layer of material of high refractive index (HI) has a physical thickness ranging from 10 to 200 nm.
65. (Previously presented) The article of claim 64, wherein the layer of material of high refractive index (HI) has a physical thickness ranging from 80 to 150 nm.
66. (Previously presented) The article of claim 65, wherein the layer of material of low refractive index (LI) has a physical thickness ranging from 40 to 150 nm.
67. (Previously presented) The article of claim 66, wherein the layer of material of low refractive index (LI) has a physical thickness of 90 nm.
68. (Previously presented) The article of claim 44, wherein the organic matrix of the composition (HI) is a hydrolysate of an epoxyalkoxysilane.
69. (Previously presented) The article of claim 68, wherein the epoxyalkoxysilane contains an epoxy group and three alkoxy groups, these latter being directly linked to the silicon atom.
70. (Currently amended) The article of claim 69, wherein the epoxyalkoxysilane corresponds to the formula (I):



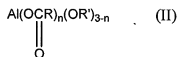
in which:

- R<sup>1</sup> is an alkyl group of 1 to 6 carbon atoms,
- R<sup>2</sup> is a methyl group or a hydrogen atom,
- a is an integer between 1 and 6,
- b represents  $\llbracket 0,1 \rrbracket$ , 0, 1 or 2.

71. (Previously presented) The article of claim 70, wherein R<sup>1</sup> is a methyl or ethyl group.
72. (Previously presented) The article of claim 70, wherein the epoxyalkoxysilane is  $\gamma$ -glycidoxypolytrimethoxysilane.
73. (Currently amended) The article of claim 44, wherein the hardenable composition of the high index layer (HI) is combined with a catalyst constituted of an aluminum compound selected from:

aluminum chelates,

compounds of formula (II) or (III):



in which:

R and R' are linear or branched chain alkyl groups of 1 to 10 carbon atoms,

R'' is a linear or branched chain alkyl group of 1 to 10 carbon atoms, a phenyl group, or a group



in which R has the meaning specified above, and n is an integer from 1 to 3, and an organic solvent, the boiling point T of which, at atmospheric pressure, ranges from 70°C to 140°C, being present in the hardenable composition (HI) when the catalyst is an aluminum chelate.

74. (Previously presented) The article of claim 73, wherein the catalyst of the hardenable composition (HI) is an aluminum chelate.
75. (Previously presented) The article of claim 73, wherein the catalyst of the hardenable composition (HI) is an aluminum acetylacetonate.
76. (Previously presented) The article of claim 44, wherein the precursor compound (I) of the second hardenable composition (LI) is a tetraalkoxysilane.

77. (Previously presented) The article of claim 44, wherein the precursor compound (I) of the second hardenable composition (LI) is a tetrathoxysilane.
78. (Previously presented) The article of claim 44, wherein precursor silane (II) is selected from perfluorosilanes.
79. (Previously presented) The article of claim 44, wherein the substrate is a substrate made of an organic glass.
80. (Previously presented) The article of claim 79, further comprising an anti-abrasion coating and/or an impact-resistant coating.
81. (Previously presented) The article of claim 44, further comprising a hydrophobic anti-fouling coating deposited onto the anti-reflection coating.
82. (Currently amended) A process for the manufacture of an article according to claim 44, comprising:

depositing onto at least one of the surfaces of the substrate optionally coated with an anti-abrasion coating or a primer layer and an anti-abrasion coating at least one layer of material of high refractive index (HI), by application and then hardening of a first hardenable composition (HI) comprising an organic-inorganic hybrid matrix resulting from the hydrolysis and condensation of at least one precursor compound bearing an epoxy or (meth)acryloxy group and at least two functions hydrolysable to silanol groups, within which at least one metal oxide and/or at least one chalcogenide is dispersed in the form of particles having a diameter of 1 to 100 nm; and

depositing onto said layer (HI) of at least one layer of material of low refractive index (LI), by application and then hardening of a second hardenable composition, said second hardenable composition comprising the product of hydrolysis and the condensation of:

- (i) at least one precursor compound (I) comprising four hydrolysable functions per molecule of formula



in which the groups W, identical or different, are hydrolysable groups and provided that the groups W do not all represent at the same time a hydrogen atom, and

(ii) at least one precursor silane (II) bearing at least one fluorinated group and comprising at least two hydrolysable groups per molecule,

said second composition comprising at least 10% by mass of fluorine in its theoretical dry extract (TDE), and the molar ratio  $[[I/I + II]] \frac{I}{I+II}$  of the precursor compound (I) to the sum of the precursor compound (I) + precursor silane (II) of the second composition being greater than 80%.

83. (Cancelled)

84. (Previously presented) The process of claim 82, wherein the layers of material of high refractive index (HI) and low refractive index (LI) are deposited by dip coating or spin coating.

85. (Previously presented) The process of claim 82, further comprising, between the deposition of the layer of material of high refractive index (HI) and that of the layer of material of low refractive index (LI), a surface treatment of the layer (HI) in order to prepare the surface for the deposition of the layer (LI).

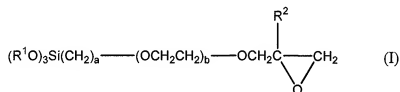
86. (Previously presented) The process of claim 85, wherein the treatment of the surface of the layer of material of high refractive index (HI) is an infrared treatment, followed by cooling by a stream of air at ambient temperature.

87. (Previously presented) The process of claim 82, wherein the anti-reflection stack is a triple layer stack (MI/HI/LI) comprising successively, and in the order starting from the substrate, a layer of material of medium refractive index (MI), a layer of material of high refractive index (HI) and a layer of material of low refractive index (LI).

88. (Previously presented) The process of claim 82, wherein the layer of material of high refractive index (HI) has a refractive index of 1.72 to 1.82.

89. (Previously presented) The process of claim 88, wherein the layer of material of high refractive index (HI) has a refractive index of 1.77.

90. (Previously presented) The process of claim 82, wherein the layer of material of low refractive index (LI) has a refractive index varying from 1.38 to 1.44.
91. (Previously presented) The process of claim 90, wherein the layer of material of low refractive index (LI) has a refractive index of 1.43.
92. (Previously presented) The process of claim 82, wherein the layer of material of high refractive index (HI) has a physical thickness ranging from 10 to 200 nm.
93. (Previously presented) The process of claim 92, wherein the layer of material of high refractive index (HI) has a physical thickness ranging from 80 to 150 nm.
94. (Previously presented) The process of claim 93, wherein the layer of material of low refractive index (LI) has a physical thickness ranging from 40 to 150 nm.
95. (Previously presented) The process of claim 94, wherein the layer of material of low refractive index (LI) has a physical thickness of 90 nm.
96. (Previously presented) The process of claim 82, wherein the precursor compound of the first hardenable composition (HI) is a hydrolysate of an epoxyalkoxysilane.
97. (Previously presented) The process of claim 96, wherein the epoxyalkoxysilane contains an epoxy group and three alkoxy groups, these latter being directly linked to the silicon atom.
98. (Currently amended) The process of claim 97, wherein the silane with an epoxy group is an epoxysilane corresponding to the formula (I):



in which:

- R<sup>1</sup> is an alkyl group of 1 to 6 carbon atoms,
  - R<sup>2</sup> is a methyl group or a hydrogen atom,
  - a is an integer between 1 and 6,
  - b represents  $\left[\left[0,1\right]\right] \underline{0}, \underline{1}$  or 2.
99. (Previously presented) The process of claim 98, wherein R<sup>1</sup> is a methyl or ethyl group.

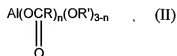


100. (Previously presented) The process of claim 98, wherein the epoxysilane is  $\gamma$ -glycidoxypropyltrimethoxysilane.

101. (Currently amended) The process of claim 82, wherein the hardenable composition (HI) is combined with a catalyst constituted by an aluminum compound selected from:

aluminum chelates,

compounds of formula (II) or (III):



in which:

R and R' are linear or branched chain alkyl groups of 1 to 10 carbon atoms,

R'' is a linear or branched chain alkyl group of 1 to 10 carbon atoms, a phenyl group, or a group



in which R has the meaning specified above, and n is an integer from 1 to 3, and an organic solvent, the boiling point T of which, at atmospheric pressure, ranges from 70°C to 140°C, being present in the hardenable composition (HI) when the catalyst is an aluminum chelate.

102. (Previously presented) The process of claim 101, wherein the catalyst of the composition (HI) is an aluminum chelate.

103. (Previously presented) The process of claim 101, wherein the catalyst of the composition (HI) is an aluminum acetylacetonate.

104. (Previously presented) The process of claim 82, wherein the precursor compound (I) of the composition of low index (LI) is a tetraalkoxysilane.

105. (Previously presented) The process of claim 82, wherein the precursor compound (I) of the composition of low index (LI) is a tetraethoxysilane.

106. (Previously presented) The process of claim 82, wherein the precursor silane (II) of the composition of low index (LI) is a perfluorosilane.